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DECONTAMINATION OF RADIOACTIVE WASTE AIR. I

by

R. Philip Hammond

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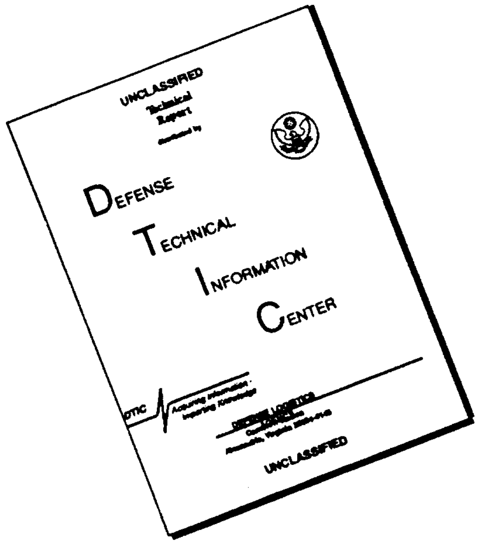

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DECONTAMINATION OF RADIOACTIVE WASTE AIR. I

R. Philip Hammond

SUMMARY

Contaminated waste air has been treated with a baffle-plate scrubbing tower with 42 plates in three stages. The air was sampled for activity before and after each stage and the effect of several process variables investigated. With only six plates per stage in use the efficiency for plain cold water washing was 99.6 per cent or better, and was 99.92 per cent for the "cloud chamber" mode of operation. Some data on particle size and preliminary engineering figures are given.

1. INTRODUCTION

The problem of disposal of contaminated waste ventilation air from laboratory and plant operations with radioactive materials has been studied as a chemical engineering operation. A survey of methods of air treatment showed that wet collection offered several advantages over dry methods when radioactive contamination is involved. Some of these advantages may be listed as follows:

1. The amount of material to be collected from the air stream is always very small in actual mass when compared with direct liquid wastes from the same operations. This means that a concentrated suspension, solution, or sludge containing the material removed from the air will be an infinitesimal addition to the liquid wastes which have to be handled in any case.

2. In case direct burial is the indicated disposal means, the concentrated sludge from a wet process will have smaller volume than the plugged filter element from a fibrous filtration system unless the filter medium is itself combustible.

3. If the filter medium is to be burned the stack gas problem occurs all over again, and it is much like disposing of a pile of dirt by digging a hole for it.

4. In the case of alpha-active contamination, disposal of a dry residue presents a considerable health problem during handling, in order to prevent redispersal of the contamination. A wet residue, on the other hand, may be pumped to the final disposal area or handled in several ways without fear of redispersal.

5. In the case of beta-gamma contamination, a dry filtration system usually stores and concentrates the collected activity in one place, so that for even moderately high level work the filter medium becomes too "hot" to approach, and must be replaced, transported, and disposed of by completely remote-operated and fool-proof mechanisms of high cost. Most wet collection systems, however, do not store the contamination removed from the air, but discharge it continuously from the system, thus preventing a build-up of activity.

6. In the case of handling air streams containing moisture, fumes, acids, and other corrosives in addition to radioactive contamination, many dry filtration systems will fail completely unless extreme dilution is provided, whereas a wet system may be easily controlled to the proper pH to prevent corrosion.

7. A wet collection system will operate at constant efficiency, while a dry filtration method gradually becomes plugged, develops leaks, and requires increased blower horsepower until the filter element is changed. It is hardly practical for cost reasons to provide continuous mechanical replacement of filter element.

A. Previous Work. Other work on wet collection has been largely limited to industrial dust and fume treatment, where high concentrations of material are present in the air. It was judged that performance of such equipment could not be predicted for the minute loadings encountered in the usual waste gas from laboratories handling radioactivity. Other workers[1] had studied collection of radioactive aerosols in wet scrubbers of the packed column type, using Raschig rings or Berl saddles. The results were not favorable in these tests since only short packed sections could be used without exceeding allowable pressure drops.

B. Type of Wet Collector. Experiments with packed columns as mentioned above would be expected to give good results only on soluble gases or large particulates. For the most important size range in radioactive aerosols (.1 to 5 μ) one would expect poor efficiencies since a packed column depends on diffusion, and both gas and liquid flows are largely in the viscous range.

Other types of wet collectors, however, do not suffer from the limitations of pressure drop, viscous flow, and low capacity exhibited by packed columns. Such types are spray towers, capillary air washers, baffle-plate scrubbers, and various contactors such as "Rotoclones" and Venturi scrubbers. After consideration of several factors such as cost of construction, pressure drop, possibility of multiple contacting, probable efficiency, etc., the simple baffle-plate tower was chosen for experimental testing on radioactive aerosols.

2. DESCRIPTION OF APPARATUS AND METHOD

A. Construction. The pilot plant now in operation consists of a baffle-plate tower of 42 plates, divided into three sections or stages in series, with a separate water circulation system for each. Each of the three towers or stages is identical in construction except that tower No. 1 is equipped with a Lucite front for observation. The towers are 15″ × 18″ × 96″ in inside dimensions and are made of redwood. They stand on 55-gal drums which act as reservoirs (Fig. 1).

The alternately opposed baffle plates have an opening area of 0.73 square feet per plate. Water in each 55-gal reservoir is circulated continuously by means of a centrifugal pump to the topmost plate of the stage, whence it cascades over the 14 plates before returning to the reservoir. An alternative discharge arrangement permits admission of the water onto the sixth plate instead of the 14th. Heat exchangers are provided by means of which any water stream may be heated or cooled.

B. Operation. The air to be treated is drawn upward through each tower in turn in countercurrent flow to the water by a blower located at the end of the system. At the top of each tower is an entrainment eliminator. These eliminators must work efficiently or the air sample taken behind it will contain activity from the recirculated water as well as from the air stream. Air pressure is measured at the top and bottom of each tower as are the temperatures of air and water streams. These instruments, together with water and air flow meters and controls, are shown in Fig. 1.

C. Sampling. The air stream is sampled as it enters the apparatus and as it leaves each of the three stages, making four samples in all. A sample which represents from 1 to 5 per cent of the air in the duct is passed through a preheater (100°F) and filtered on 25 sq in. of H-70 paper.* It then passes through a calibrated orifice flow meter and control valve and is exhausted by an auxiliary blower. The sample is not returned to the system. The preheater prevents the deposition of free water on the filter paper, which would reduce the air flow and change the filtering characteristics (Fig. 2).

D. Counting of Samples. The four filter papers from each run are counted for alpha activity with a counter having a large multi-wire probe which will cover the entire 4 by 9-in. area of the paper. (A thin paper mask is used in the filter holder to restrict deposition of the activity to a 25-in.^{2} area.) By comparison with a standard alpha sample each count is corrected to 50 per cent geometry. The counting rate is determined to a 9/10 error of 2 per cent. From the time and flow-rate used in collecting the sample the concentration of radioactivity in the gas stream may be computed. For the

*Hollingsworth-Vose Co.

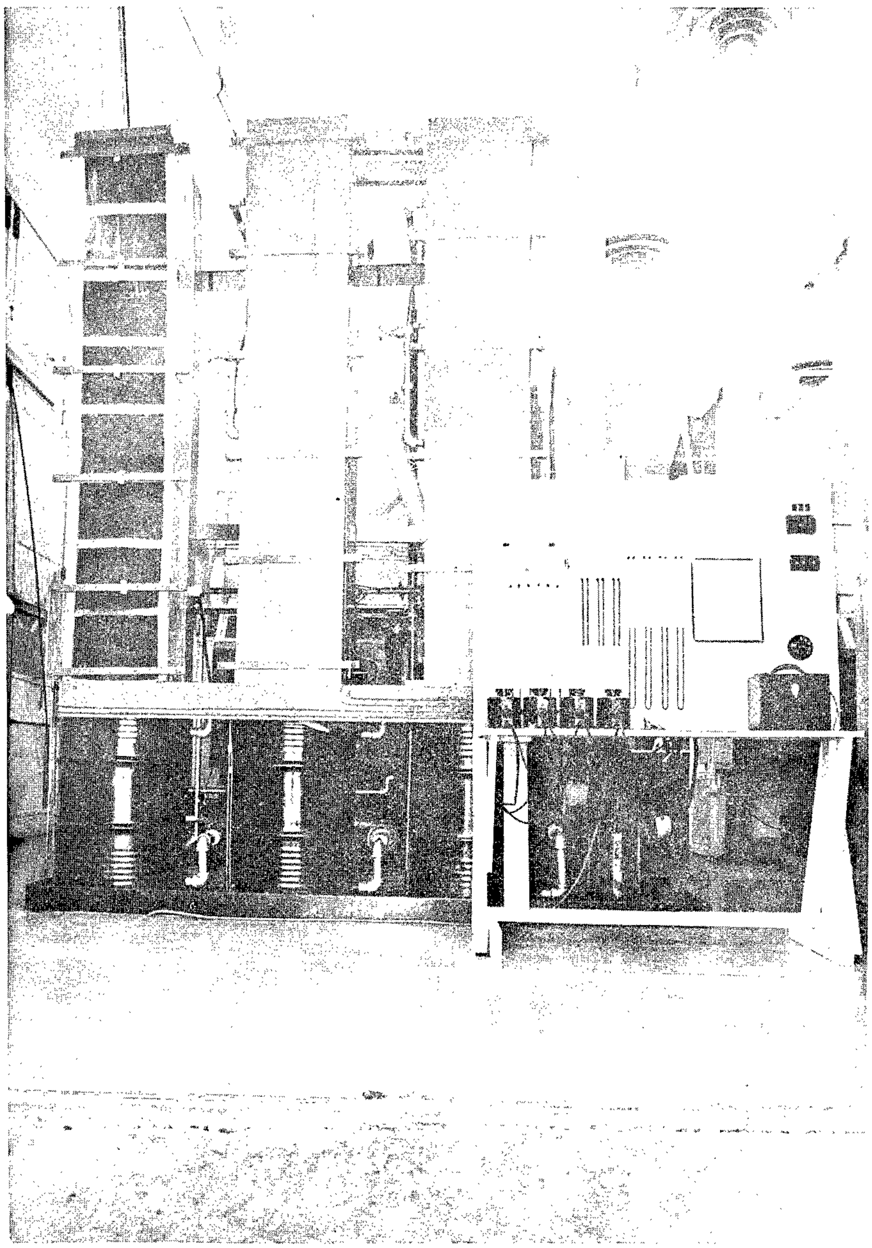

Fig. 1—Front view of pilot plant.

present work the empirical concentration unit of counts per minute per cubic foot has been used. Concentrations encountered in this work have ranged from 1 to 20,000 c/m/ft^3. Such concentrations correspond to about 4×10^{-6} grains/ft^3 as compared to ordinary industrial dust loadings of about 0.3 grains/ft^3.

3. VARIABLES STUDIED. RESULTS

A total of over 100 runs has been made to date, with the aim of studying the effect of certain variables on the collection efficiency of the apparatus. As much as possible all conditions were held constant except the variable being investigated.

A. Feed Concentration. The contaminated air used for all runs was drawn from an exhaust duct serving a factory processing operation in which a dry, insoluble salt of an alpha emitter is handled. On certain days the operation being done resulted a higher dust concentration than on other days. On days of high dust concentration it is believed that the main contamination was released in large bursts, but no control or measurement of this was possible, since the sample paper recorded only the total contamination received in the duration of the run (2 to 4 hr). Thus the effect of feed concentration could only be studied by duplication of runs on different days. To date no significant variation in efficiency with feed concentration has been detected in the range of 1 to 20,000 c/m/ft^3.

B. Air Rate. Table I indicates that no decrease in overall efficiency is encountered in going from 80 to 160 cfm (air velocity, 110 to 220 ft/min). The average removal was 99.60 per cent at 80 cfm and 99.70 per cent at 160 cfm. Still higher air rates are presently under investigation, but indications are that pressure drop increase will limit this variable before loss of efficiency is encountered. This would indicate that contact time is not a significant variable in the range used.

Table I—Effect of Increasing Air Rate from 80 cfm to 160 cfm*

Run No.	Air rate, cfm	Overall removal, %	Contamination level c/m/cu ft. Feed	Output
57	80	99.99	3.03	0.00
58	80	99.69	13.7	0.42
59	80	98.93	6040	64.8
77	80	99.53	7.5	0.03
78	80	99.86	25.2	0.03
		Average 99.60		
53	160	99.69	1206	3.75
60	160	99.53	2260	10.40
61	160	99.48	3310	17.20
62	160	99.99	63.4	0.00
63	160	99.92	96.8	0.07
64	160	99.70	417	1.26
65	160	99.99	73.8	0.00
66	160	99.32	201	1.37
		Average 99.70		

*Six plates per tower in use.

C. Water Rate. Table II summarized the results of groups of runs at different water rates as compared to the efficiency of tower No. 1, other conditions being held constant. The effect is apparently not great as long as the water rates are sufficient to maintain a continuous film across the edge of the baffle plate (about 10 gpm for a 15-in. shelf, or 7 gpm per ft of shelf edge).

Table II—Effect of Varying Water Rate in Tower No. 1*

Water rate, gpm	Tower No. 1 efficiency (average), %	Pressure drop per plate, in. of water
10	76.0	0.021
15	76.4	0.025
19	86.7	0.028
31	82.7	0.036

*Air rate 160 cfm.

It should be noted that increased water flow causes increased pressure drop, so that a slightly reduced water rate with a slightly decreased efficiency per plate might result in a superior design having more plates but a lower pressure drop.

D. Hot Water Washing. Column 2 of Table III shows the effect of using 95°F water in tower No. 2 as compared with cold (63°F) water. The significant increase in efficiency from 82 per cent to 92 per cent may be an indication of better wetting action. The use of wetting agents will be tried.

Table III—Comparison of Plain Cold Water Washes with 95°F Cloud-Chamber Mode of Operation, Other Conditions Held Constant*

	Removal efficiency, %			Contamination level c/m/cu ft.	
Run No.	Tower No. 2	Tower No. 3	Overall	Feed	Output
53 (cold water)	78	83	99.69	1206	3.75
60 (cold water)	85	84	99.53	2260	10.40
61 (cold water)	78	88	99.48	3310	17.20
62 (cold water)	96	99	99.99	63.4	0.00
63 (cold water)	87	92	99.92	96.8	0.07
64 (cold water)	79	90	99.70	417	1.26
65 (cold water)	78	99	99.99	73.8	0.00
66 (cold water)	79	80	99.32	201	1.37
Average	82	89	99.70		
50 (hot water)	97	45†	99.95	140.6	0.07
73 (hot water	97	81	99.90	24.4	0.03
74 (hot water)	98	49†	99.84	37.1	0.06
75 (hot water)	94	88	99.97	949	0.24
79 (hot water)	93	98	99.92	56.4	0.04
80 (hot water)	68	97	99.84	3940	6.21
81 (hot water)	98	99	99.99	129	0.00
82 (hot water)	96	99	99.99	234	0.00
Average	92	94	99.92		

*Six plates per tower in use.
†Values not included in average.

E. "Cloud Chamber" Effect. When runs were made with tower No. 2 at elevated temperatures, tower No. 3 was cooled with a heat exchanger to 65 to 73°F, thus removing the heat added in tower No. 2, and causing "fogging" or condensation of the excess humidity produced. This condensation

should occur preferentially on the radioactive particles as nuclei (within certain size ranges) and thus increase their particle size and ease of collection. The results may be noted in column 3 of Table III.

Table IV shows overall efficiency for "cloud chamber" runs with tower No. 2 at 110°F.

Table IV—Cloud Chamber Runs at 110°F*

Run No.	Overall removal efficiency, %	Contamination level c/m/cu ft. Feed	Output
45	99.95	6860	3.28
46	99.99	1262	0.00
47	99.99	350	0.00
48	99.98	10650	1.70
49	99.68	29.7	0.09
95	99.99	254	0.00
96	99.89	252	0.28
97A	99.99	314	0.01
97B	99.98	179	0.03
98A	99.62	53.1	0.20
98B	99.99	2070	0.00
98C	99.98	3460	0.83
99	99.98	228	0.05
100	99.99	841	0.00
101	99.99	161	0.00
102A	99.99	861	0.01
102B	99.99	531	0.10
103A	99.95	1970	0.87
103B	99.99	8230	0.72
104	99.99	142	0.00
	Average 99.95		

*Six plates per tower in use.

F. Effect of Fresh vs. Contaminated Water. The water in the circulating systems has been used for weeks and even months of daily runs without apparent ill effect on the results obtained. (Evaporation losses were made up by fresh water.) When the reservoirs were drained, flushed, and refilled, no apparent improvement in results was obtained. It should be noted, however, that the radioactive salt present is insoluble and forms partly a suspension and partly a sludge in the reservoirs. The suspensions formed reached concentrations as high as 1 microcurie per liter.

4. PARTICLE-SIZE MEASUREMENTS

The concentrations of radioactive dust encountered in the work were so minute that the most desirable method of size testing, thermal precipitation, could not be used. As a substitute a radioautographic technique was developed by one of the authors. This technique is described in a separate report, LAMS-905, by J. A. Leary. The results of this analysis are shown in Fig. 4. Curves 2A and 2B indicate that tower No. 1 collects essentially all particles larger than 2.7 μ in diameter, while curve 3A shows that tower No. 3 passes none above 1.0 μ. This undoubtedly represents the higher probability of collection of large particles, and serves as a warning that the efficiencies reported here would only apply to material having size distributions represented by curves 1A and 1B.

5. DISCUSSION OF RESULTS

A. Sources of Error. 1. Sampling errors. The sampling flow-rate orifices were calibrated to the accuracy with which the manometers could be read, about 5 per cent. For best results the velocity of air in the sampling line should be the same as in the duct being sampled. With the apparatus used, this was possible only at the lower air rates. The sample lines for higher air rates operated at a lower velocity than the ducts, which conceivably might have allowed large particles to settle out and fines to bypass the sampling throat.

2. Counting errors. The counts were carried out to provide a statistical error of about 2 per cent with 90 per cent certainty. Other errors bring the overall errors to about 4 per cent. Combining this error with that in sample flow-rate, it can be calculated that the overall error is 0.08 per cent in a true overall efficiency of 99.0 per cent becoming less than this for better efficiencies.

3. Filter paper errors. There is undoubtedly some penetration of the filter paper by the finer particles, which would not then be counted. Tests were run in which two filter papers were used in the holder during a highly contaminated run. The front and back of each paper was counted to estimate the penetration of the paper by fine particles. The proportion of particles able to completely penetrate the first paper was found to be negligible. The assumption has been made in this work that such errors affect all samples equally.

4. Entrainment. One of the most serious sources of error is in faulty entrainment elimination. Since the water is recirculated and attains very high contamination levels with respect to the air it is cleaning, even a few drops of entrained liquid carried over to the preheater and filter would deposit significant amounts of activity in addition to that actually present in the gas stream. This error, however, is a conservative error in that it tends to lower the observed collection efficiency. The installation of improved eliminators may therefore show that the towers are operating at better efficiencies than now indicated.

5. Contamination of samples. A source of error which is difficult to measure or predict is the contamination of the filter paper by activity remaining in the duct or on the sample holder from a previous run. This error would affect mostly the final output sample, which is often only a few counts above background, and would be in the direction to cause a lower apparent efficiency.

B. Preliminary Engineering Study. It having been established that baffle-plate towers are capable of suitably high efficiencies for consideration, it was desired to compare the apparatus required for this method with other methods on an economic basis. Such a study is now underway. Structural details, power, water, and heat consumption, will be determined. Preliminary considerations lead to the following observations:

1. The first cost of a complete installation including blower and high velocity stack should not exceed $2.00 per cfm for an installation of 20,000 cfm or more. Further improvements in design may reduce this considerably. The apparatus is of a type which need not be housed.

2. Water consumption should not greatly exceed that required to saturate the air at the discharge conditions. It is presently planned to use a scrubber installation as evaporators for the contaminated liquid waste from laboratory operations. The waste will be neutralized and used as make up to the towers, with only sufficient blow-down to prevent salt incrustation. Such a system thus treats both liquid and gaseous wastes from laboratory operations with radioactivity and discharges a single concentrated waste stream in easily handled form.

3. The maximum air velocity investigated so far is 330 ft/min. At this rate the pressure drop per plate at an average water flow is about 0.025 in. of water. The total pressure drop for a complete system of 45 plates with three sets of eliminators may thus be in the neighborhood of 2.5 to 3 in. of water. The floor space required would be approximately 8 square feet per 1000 cfm. The blower would be mounted on top of the tower at a height of about 30 ft, and would deliver into a short high-velocity stack.

4. Recirculation of water will require approximately 1 kw per 1000 cfm if three separate recirculation stages are used. At periods of light load it would be possible to shut down one or more stages.

REFERENCE

1. Report HDC-611, Interim Report, by C. E. Lapple, Hanford Engineer Works.

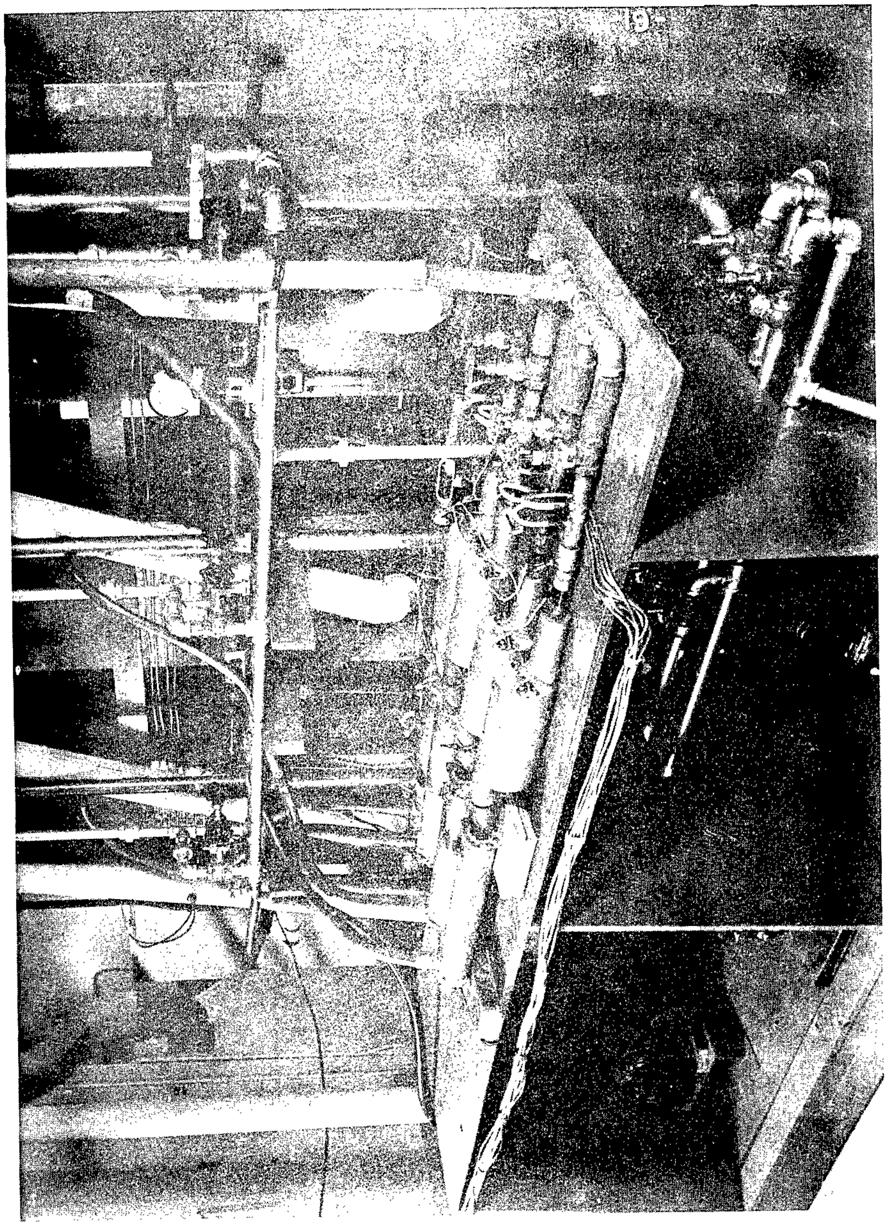

Fig. 2—Detail of sampling equipment.

Fig. 3 — Scrubbing action in tower.

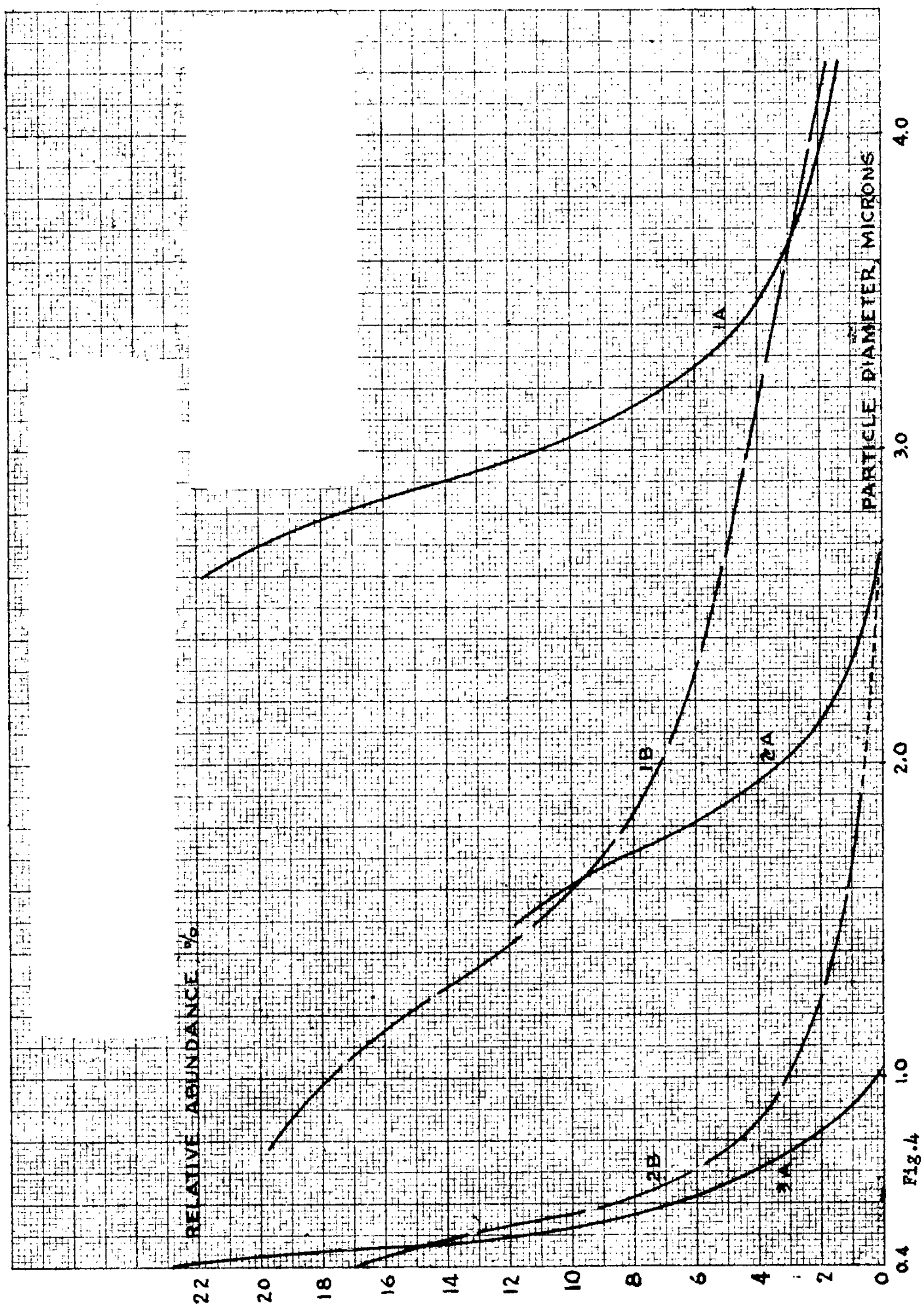

Fig. 4 — Particle size distribution in feed to and discharge from system. For particles greater than 0.4 μ. 1A, Feed to tower No. 1 (dumping); Run A. 2A, Discharge from tower No. 1; Run A. 3A, Discharge from tower No. 3; Run A. 1B, Feed to tower No. 1 (no dumping); Run B. 2B, Discharge from tower No. 1; Run B.

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